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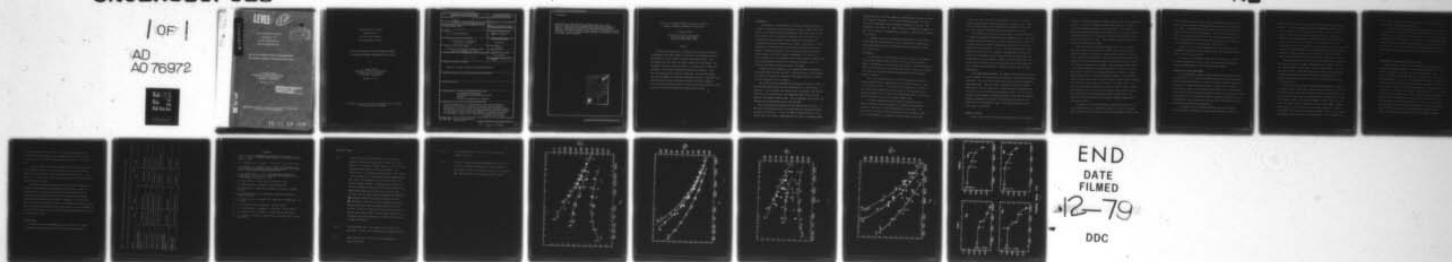
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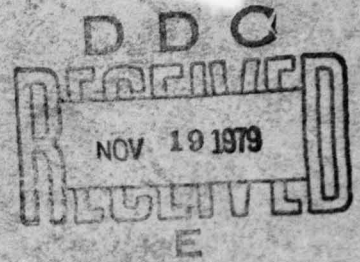
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Technical Report on Research

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MORE ON THE ACOUSTICAL METHOD FOR MEASURING ENERGY
AND TANGENTIAL MOMENTUM ACCOMMODATION COEFFICIENTS

by

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The University of Mississippi
University, Mississippi

November 1, 1979

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More on the Acoustical Method of Measuring Energy and
Tangential Momentum Accommodation Coefficients

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Abstract

Additional measurements of the energy (EAC) and tangential momentum accommodation coefficients (TMAC) of He and Ne on polycrystalline tungsten have been made using an acoustic technique previously reported. The measurements were made after the tungsten surface had been exposed to the atmosphere and after it had been flashed in a vacuum at 1240, 1460, 1770 and 2000°K. Three stable, reproducible surfaces were detected. These have been associated with an atomic oxygen film, upper molecular oxygen film, and an unknown film produced by atmospheric exposure. Comparison with molecular beam scattering experiments indicates that TMAC corresponds to the fraction of molecules scattered diffusely from polycrystalline surfaces and EAC to the diffuse fraction from single crystal surfaces.

INTRODUCTION

The exchange of energy between a solid surface and gaseous molecules striking it traditionally has been measured by using a thermal conductivity cell.¹ In this method the energy accommodation coefficient (EAC) is determined from the rate at which heat is conducted from a wire exposed to a rarified gas. A second method involves measuring the distribution of scattered particles when a molecular beam strikes a solid surface. In this experiment, the reflected beam is resolved into specular and diffuse components. At least from single crystal surfaces, the diffuse component is usually assumed to have been thermally accommodated.² Of course, the direct verification of this assumption would involve measuring simultaneously the scattering angle and energy of the scattered particles. This is a difficult experiment to perform and results are meager.³

Relatively few measurements of the tangential momentum accommodation coefficient (TMAC) have been reported.⁴ Practically none of the experiments that have been reported have attempted to determine the effect of adsorbed surface contaminants upon this quantity.

The acoustic method for measuring EAC and TMAC was reported in 1975⁵. It involves measuring the sound velocity and absorption in a gas confined to a small tube. At low pressure, the sound velocity and absorption are sensitive functions of the "slip velocity" and "temperature jump" at the tube wall. These, in turn, depend on TMAC and EAC. Thus, the measured sound velocity and absorption yield simultaneous values of EAC and TMAC.

One can postulate two situations in which TMAC and EAC can differ. In the first, the "diffuse fraction" can contain molecules that are scattered elastically but diffusely. In this case TMAC is greater than EAC. In the second, some of the energy accommodated molecules could be exchanging energy

preferentially with the normal component of momentum while leaving the tangential component unchanged. In this case, EAC would be greater than TMAC. The acoustic measurements have been analyzed assuming that the first case (TMAC > EAC) is the likely one. TMAC is found to be significantly larger than EAC. Their difference is interpreted as a measure of the molecular roughness of the surface.

In comparing the acoustic and molecular beam results, a correlation is found between EAC and the diffuse fraction scattered from a single crystal surface and between TMAC and the diffused fraction scattered from a polycrystalline surface.

EXPERIMENTAL

The experimental system has been previously described.⁵ The tungsten tube and sound transducer were the same as in the previous experiments. However, the diaphragm in the sound source was replaced. Flashing temperatures of the tube were measured more accurately than before. Emissivity and window corrections for the optical pyrometer readings were taken from National Bureau of Standards.⁶

One of the unexpected results of the 1974 measurements was that the flashed tungsten surface remained stable over a period of weeks in a vacuum of the order of 10^{-8} torr. The stability of the surface in this relatively poor vacuum was attributed to the effectiveness of the mischmetal getter that was evaporated onto the walls of the vacuum system and to the gettering action of the outer surface of the flashed tungsten tube.

The present experiments further tested the effectiveness of the getter in removing surface contaminants at relatively high background pressures. This time, the system was baked out at only 150°C. If a sufficiently clean system could be obtained with this low bake out temperature, the diaphragms in the transducers would be subject to much less

thermal stress and, therefore, be less subject to failure. The lowest pressure attained after evaporating the getter was about 1×10^{-7} . However, the inside surface of the tungsten tube remained stable for three days after flashing at 2000°K even in this relatively poor vacuum.

The tungsten tube was made by Ultramet Company by chemical deposition of tungsten from tungsten fluoride gas on a stainless steel rod. Impurities, including carbon, were estimated to be at most a few parts per million. The stainless surface upon which the tungsten was deposited was specified as having a 10 microinch finish. (i.e., 0.7 times the average peak to valley difference is equal 10 μ in.) According to the manufacturer, the crystal growth in the deposition process was such that the 110 crystal plane was perpendicular to the tube surface. However, the inner surface, which was in contact with the stainless rod in the deposition process, was expected to have a random crystal orientation. This was the surface that was under investigation.

In the experimental procedure, the tungsten tube which had been exposed to the atmosphere was heated briefly by induction to the specified flashing temperature. The tube was then cooled and the sound velocity and absorption measurements made. Then the process was repeated, raising the flashing temperature to the next higher value. Measured values of sound absorption and velocity were then compared with theoretical values. For this purpose, a computer was programmed to select the values for the energy and tangential momentum accommodation coefficients (EAC and TMAC) which gave the best theoretical fit to experimental values. The flashing temperatures were 1240, 1460, 1770 and 2000°K.

SUMMARY OF RESULTS

Figures 1 and 2 show the velocity and absorption in Ne as a function of

pressure. The parameters plotted in these figures are the real and imaginary parts of the "reduced" propagation constant as a function of r_ω , where r_ω is a reduced parameter varying as pressure/frequency.⁵ The frequency at which the measurements were made varied between 13.6 and 14.0 kHz. However, all measured values were "corrected" to 14 kHz so that the results could be plotted as a function of r_ω only. Figs. 3 and 4 are similar plots of the results of the measurements in He.

From these figures, three distinguishable states for the tungsten surface have been clearly identified. Further, these states have been repeatedly reproduced. The first of these surfaces results from exposure of the tungsten to the atmosphere. This state is not affected by the 150°C bakeout of the vacuum system and persists even when the surface is flashed in a vacuum at temperatures up to 1200°K. The second identifiable surface results from the exposure of the flashed surface to a few torr of oxygen. This surface is stable up to flashing temperatures of 1600°K. The third identifiable surface is produced by flashing at 2000°K. The EAC and TMAC values for these three states are given in Table I, and are used to calculate the theoretical curves shown in the figures for the three states. A.C. values for the surface exposed to O₂ and for the surface flashed at 2000°K were published in 1975. The recent measurements confirm the 1975 values and establish the flashing temperatures needed to produce them. A.C. values for surfaces resulting from intermediate flashing temperatures are also given in Table I.

In addition, the recent measurements establish a difference in the surface when exposed to the atmosphere and when it is exposed to oxygen. This difference had gone unnoticed in the 1974 measurements. However, when

the difference was observed in the current measurements an examination was made of some calibration measurements made as early as 1973 before the tube was flashed. This examination revealed that the same difference had been present in the 1973 and 1974 measurements. Some of the points plotted in Figs. 1-4 are from data taken in 1973, some are 1974 data and some are 1979 data. The agreement in the data taken over a period of years establishes the reproducibility of the three distinct surface states.

Results of the measurements made after the tube was flashed at the intermediate temperatures (1240, 1460, and 1770°K) are also shown in these figures plotted as solid points.

In Figure 5 the accommodation coefficients are plotted as a function of flashing temperature. The A.C. values for the three distinguishable surfaces mentioned above are also indicated in this figure.

COMPARISON WITH OTHER MEASUREMENTS

It is interesting to compare results reported here with those published by others and to try to identify the three persistent states mentioned above. Modern techniques developed in the last ten years have yielded a great amount of data on the oxygen states on the specific crystal planes of tungsten. It is difficult to compare the measurements here reported for a polycrystalline surface with these microscopic measurements for specific crystal planes. However, it is possible to see a correlation between the current measurements and polycrystalline A.C. values obtained some years ago from thermal-conductivity-cell and molecular-beam-scattering experiments.

Y.H. Wachman⁷ and P.W. Blickensderfer⁸ have made careful studies of the formation and evaporation of oxygen films on tungsten. These two

compared their results with earlier experiments going back to the nineteen thirties. In these experiments the energy accommodation coefficients of He and Ne is measured in a thermal conductivity cell. Three or possibly four different surface states of oxygen coverage are observed. Wachman and Blickensderfer, following Roberts, associate the different EAC values with an "upper molecular state", a "composite state" (partly molecular and partly atomic), and one or two "atomic states" resulting from partial and total coverage by an atomic monolayer. The state formed on the surface depends on the amount of oxygen exposure, the rate at which it is admitted to the surface and the temperature to which the surface is heated after exposure.

The upper molecular state is likely the one produced here in exposing the tube to a few torr of oxygen. Wachman measures a He EAC of 0.185 for this upper molecular film and finds it stable to about 1000°K. This would agree with our 0.18. The Ne EAC values for this state have been reported by Morrison and Roberts,⁹ Van Cleave,¹⁰ Morrison and Grummitt,¹¹ and Wachman. Values range from 0.32 to 0.41 which is within experiment error of our value of 0.45.

The surface produced with the highest flashing temperature (2000°K) is likely the atomic oxygen state. EAC for He on this surface is $0.06 \pm .03$ and is in agreement with values reported by workers referenced above. Most of them find two atomic surfaces representing different degrees of coverage by a monolayer. EAC values reported by Wachman for the two are .065 and .085. The atomic surface is found to be stable to about 1800°K. Roberts gives a value of .177 for EAC of Ne on the atomic oxygen film. Morrison and Grummitt list 0.146 and say the film is stable to 1400°. This is within experimental error of the $0.12 \pm .05$ obtained here. It is concluded that

curves c and b correspond to what earlier workers have identified as the atomic and the upper molecular oxygen films.

The surface film produced when the tube is exposed to the atmosphere is surprisingly reproducible and is clearly distinguishable from the upper molecular state of oxygen. There is nothing in the literature with which to compare the EAC and TMAC values for this state. Further experimentation is needed to identify its composition and stability. The additional coverage, whatever it is, has a striking effect on the accommodation coefficients, approximately doubling EAC and increasing the already large TMAC value by 20%.

INTERPRETING THE DIFFERENCE BETWEEN EAC AND TMAC

Molecular beam experiments usually assume that molecules scattered diffusely from single crystals are also thermally accommodated.¹² In the present experiment, this would be the same as assuming $EAC = TMAC$. For the polycrystalline surface used here, this is clearly not the case, nor should one expect it to be. For the polycrystalline surface, the diffusely scattered molecules will contain those thermally accommodated as well as those scattered elastically in random directions from the randomly oriented crystal planes. Therefore, EAC should be compared with the diffuse fraction scattered from the single crystal surface (assuming thermal accommodation of molecules scattered diffusely from this surface); and TMAC with the diffuse fraction from polycrystalline surface. This is, in fact, what is found when values obtained here are compared with the results of Weinberg and Merrill¹² for the single crystal tungsten and with the results of Ollis, Lintz, Pentenero, and Cassuto¹³ for polycrystalline tungsten.

Weinberg and Merrill (see their Fig. 8) have trapping probabilities for single crystal corresponding, within experimental error, with He, Ne and Ar EAC values for the surface flashed at 2000°C. (See ref. 5 for EAC for Ar)

On the other hand, the diffuse fraction of Ne molecules scattered from polycrystalline tungsten, as reported by Ollis, et al, agree with TMAC values for both the oxygen covered surface and the flashed surface. (See their Fig. 4. Note in this figure they have plotted directed fraction rather than the diffuse fraction.)

Of course, the molecular beam scattering fractions were measured at a single scattering angle and EAC and TMAC are average values. However, the comparison seems to confirm that the difference between polycrystal and single crystal beam scattering results is due to the elastic but random scattering of the polycrystalline surface. Furthermore, we conclude that TMAC as measured here should be the same as the fraction of molecules scattered diffusely from a polycrystalline surface, while EAC is the energy accommodation coefficient and is equal to the fraction of molecules scattered diffusely from a single crystal surface.

ACKNOWLEDGEMENT

The author gratefully acknowledges the help of Glenn Holt in making the measurements and the support of the U.S. Office of Naval Research.

TABLE I. Energy and Tangential Momentum Accommodation Coefficients for Neon and Helium on Tungsten

Surface	Neon		Helium	
	EAC	TMAC	EAC	TMAC
Exposed to atmosphere. 1973 and 1979 data combined.	0.80(± 0.10)	0.96(+.04-.06)	0.50(± 0.08)	0.92(± 0.08)
Flashed at 1240°K	0.80(+0.15-0.05)	0.96(+0.05-0.15)	0.44(± 0.10)	0.76(± 0.15)
Flashed at 1460°K	0.52(+0.15-0.07)	0.88(+0.05-0.15)	0.40(+.05-0.15)	0.72(± 1.0)
Flashed at 1770°K	0.40(+0.05-0.12)	0.72(+0.15-0.05)	0.16(± 0.05)	0.80(± 0.10 -0.20)
Flashed at 2000°K 1974 and 1979 data combined.	0.12(± 0.05)	0.53(± 0.06)	0.06(± 0.03)	0.38(± 0.05)
Flashed surface exposed to O ₂	0.45(+0.10-0.05)	0.80(+0.05-0.15)	0.22 (± 0.10)	0.75(± 0.1)

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LEGEND FOR FIGURES

- Fig. 1 Sound velocity in Ne. The abscissa, r_ω , is the reduced parameter varying as the gas pressure divided by the sound frequency. It is equal to $c_0^2 \rho_0 / \omega \eta$, where c_0 is the free space, ideal gas sound velocity; ρ_0 is the gas density; η its viscosity; and ω is 2π times the sound frequency. The ordinate is the reciprocal of the reduced sound velocity. \blacksquare measurements made, some in 1974 and some in 1979, after the tube surface had been flashed at 2000°K. \triangle measurements made, some in 1974 and some in 1979, after the tube surface had been exposed to O_2 . \circ measurements made, some in 1973, some in 1974 and some in 1979 after the tube surface had been exposed to the atmosphere. \bullet measurements made after the tube was flashed at 1240°K. \blacksquare measurements made after the tube was flashed at 1460°K. \blacktriangle measurements made after the tube was flashed at 1770°K. Curves a, b, and c are theoretical curves calculated using the TMAC and EAC values in Table I for the surface exposed to the atmosphere, the surface exposed to O_2 , and the surface flashed at 2000°K respectively.
- Fig. 2 Sound absorption in Ne. The ordinate is the reduced sound absorption. See Fig. 1 for an explanation of symbols and curves.
- Fig. 3 Sound velocity in He. See Fig. 1 for an explanation of symbols and curves.

Fig. 4 Sound absorption in He. See. Fig. 1 for an explanation of symbols and curves.

Fig. 5 Energy and tangential momentum accommodation coefficients for He and Ne on tungsten as a function of flashing temperature.

○ -- measured values for the surface exposed to the atmosphere

■ -- measured values for the surface exposed to oxygen

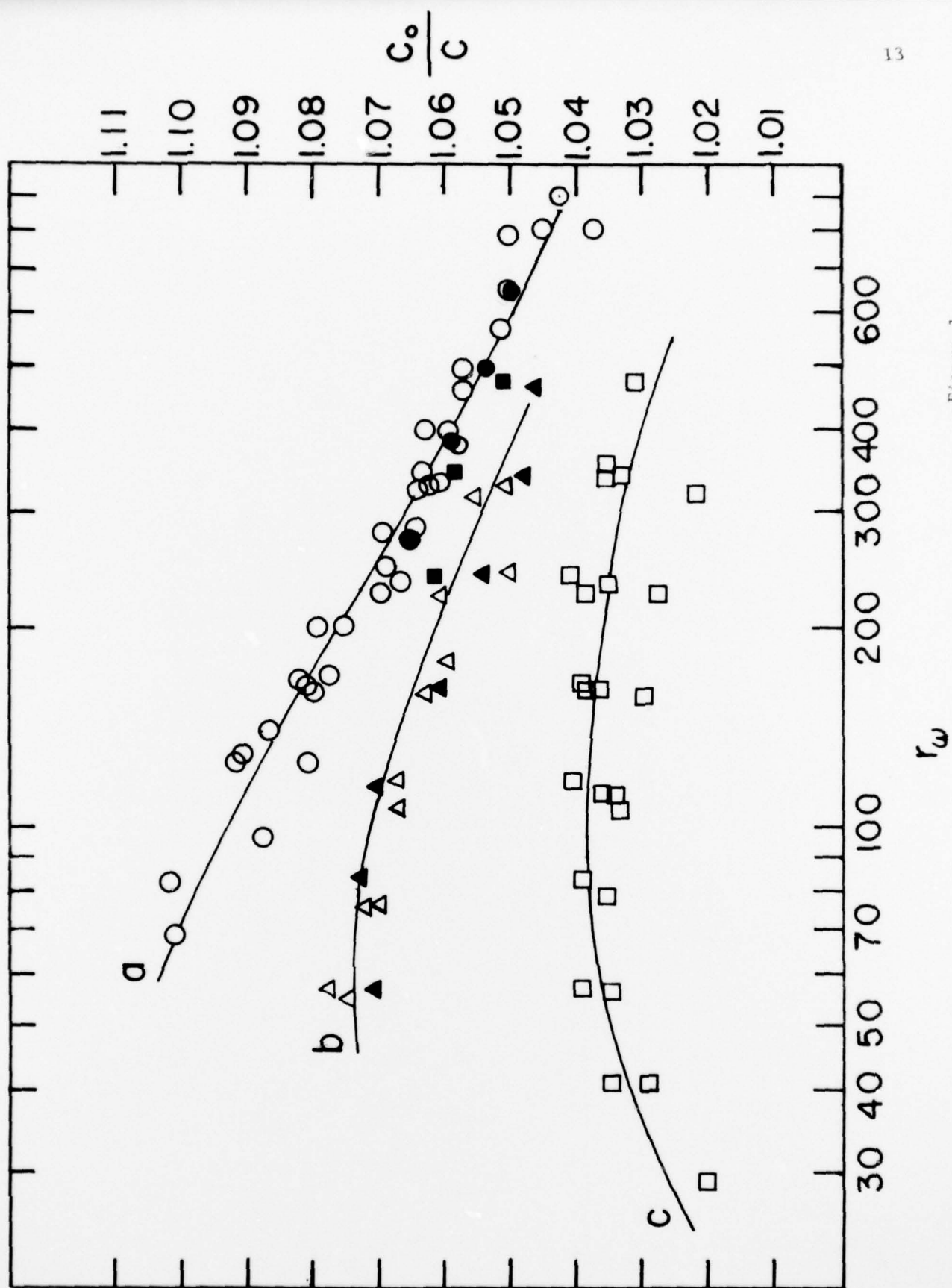


Figure 1

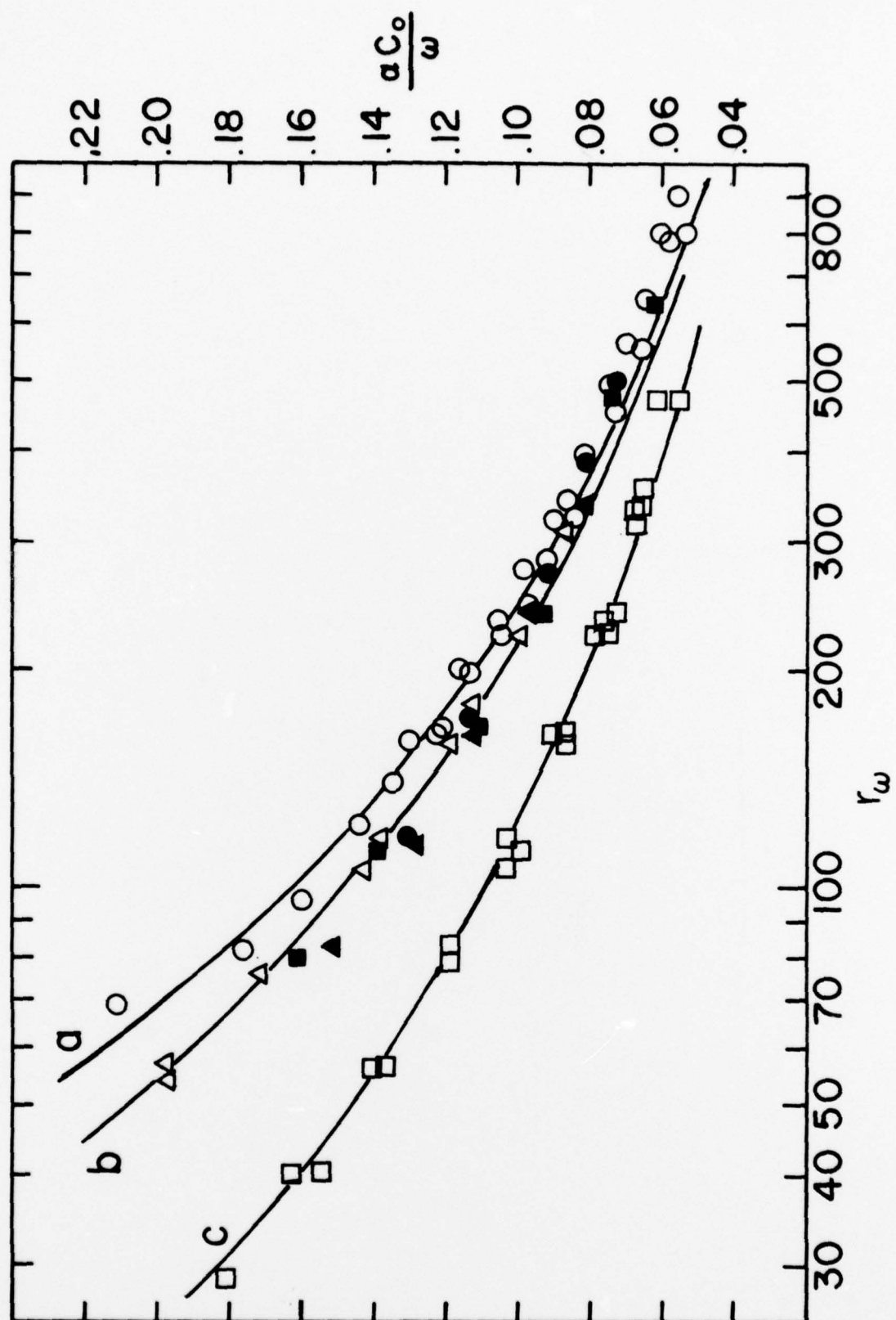


Figure 2

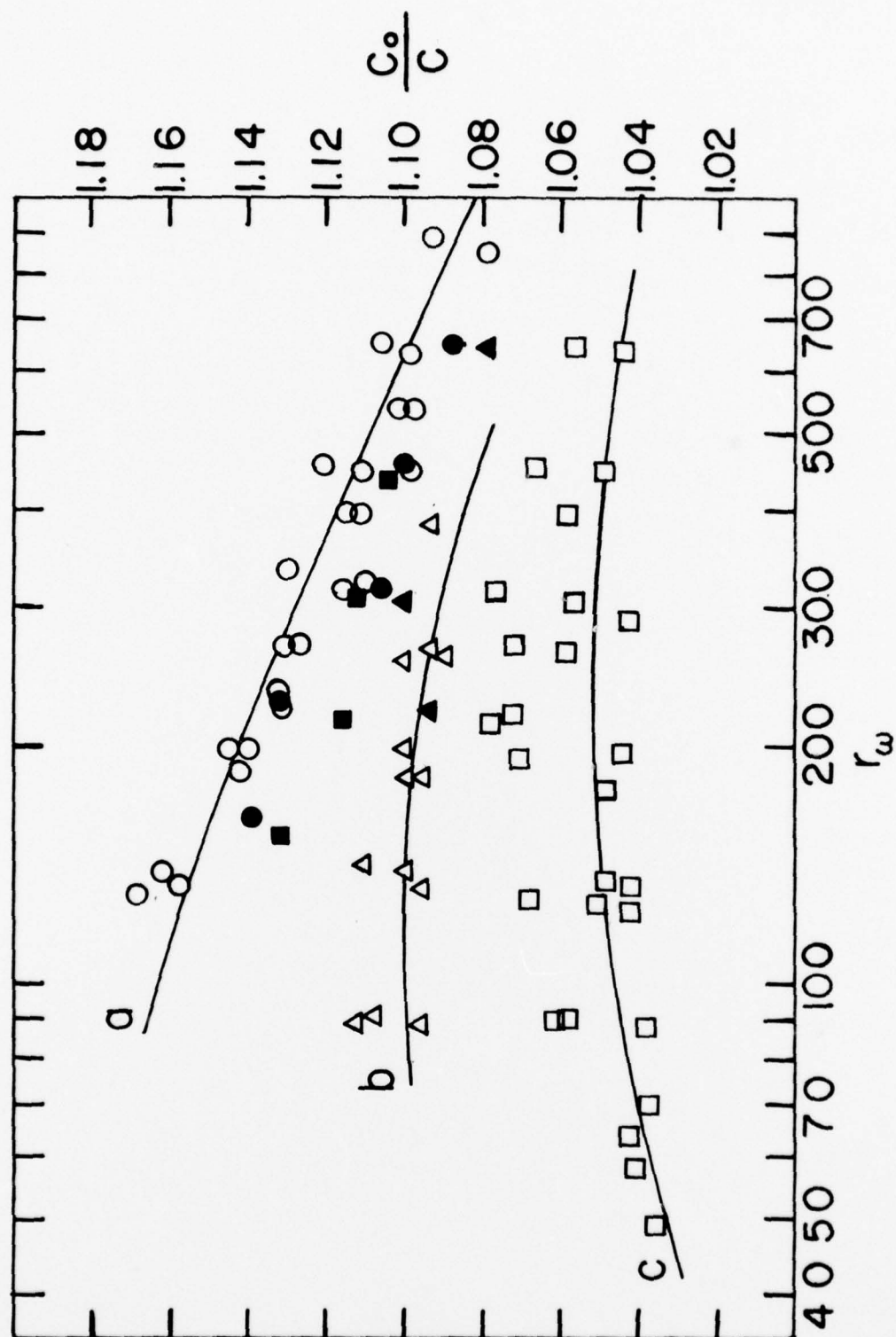


Figure 3

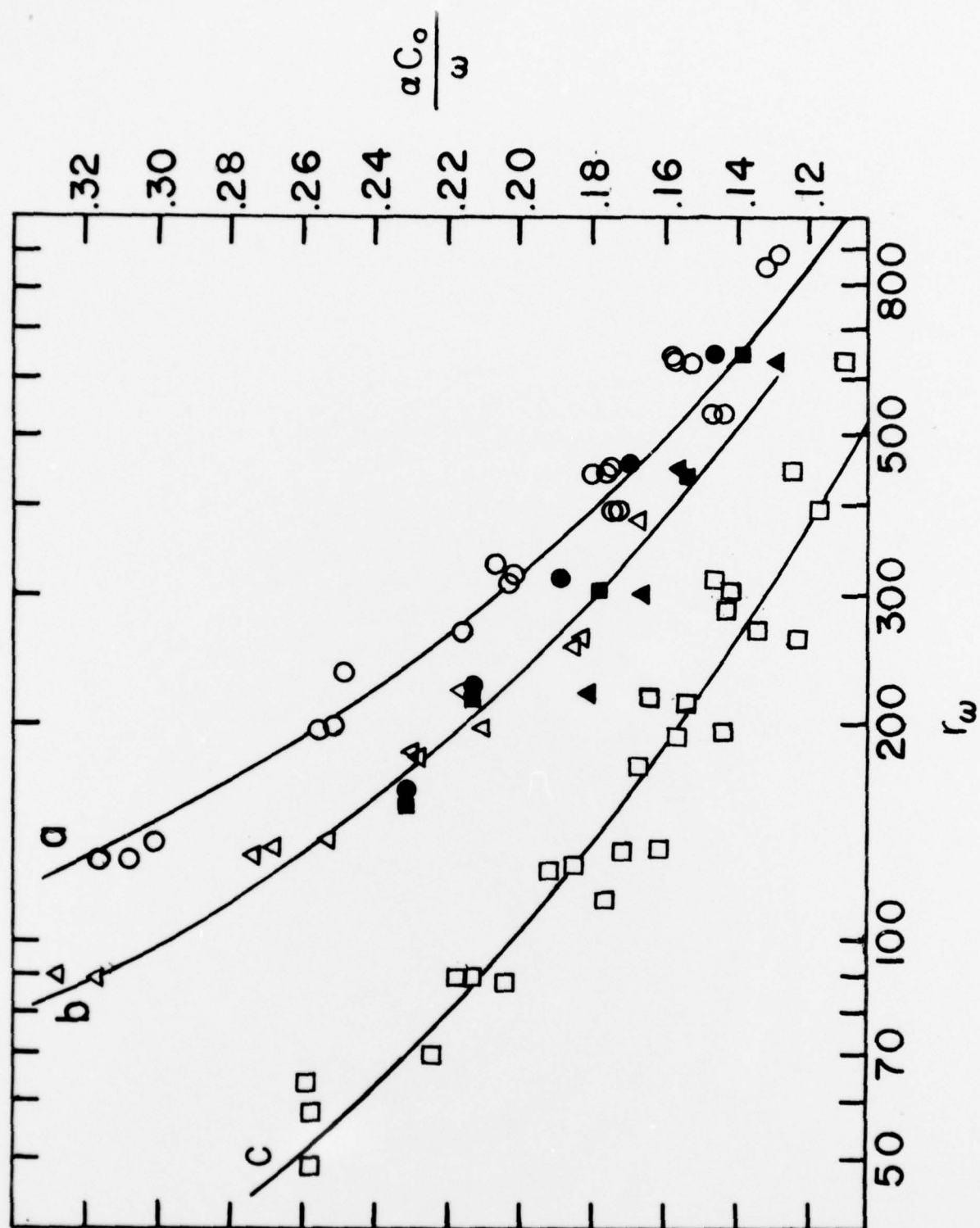
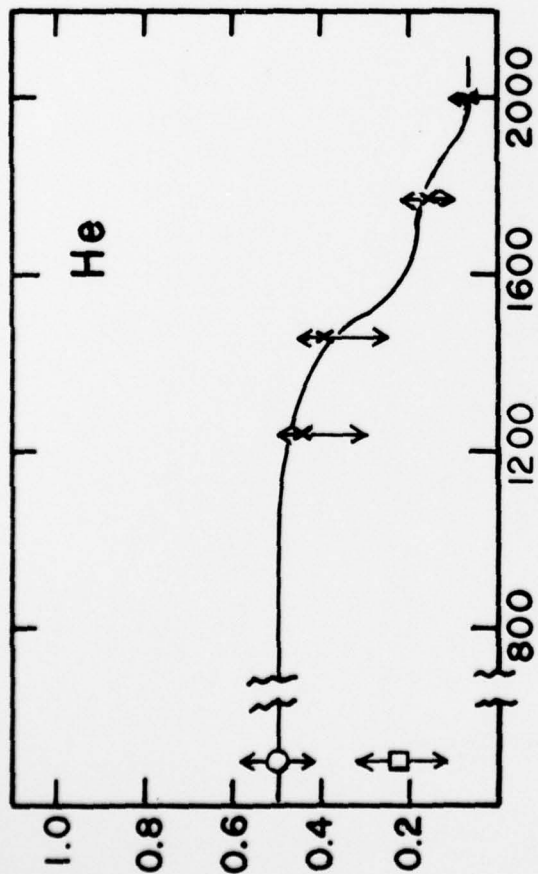
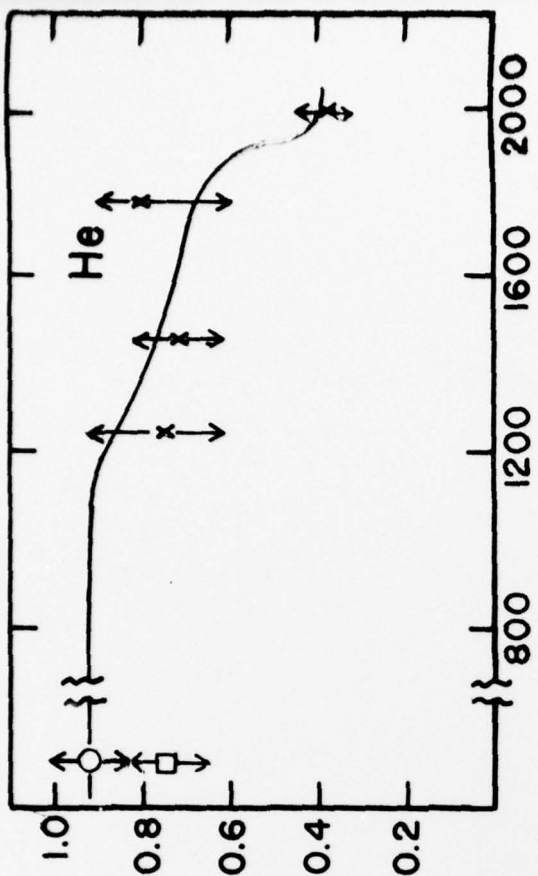


Figure 4

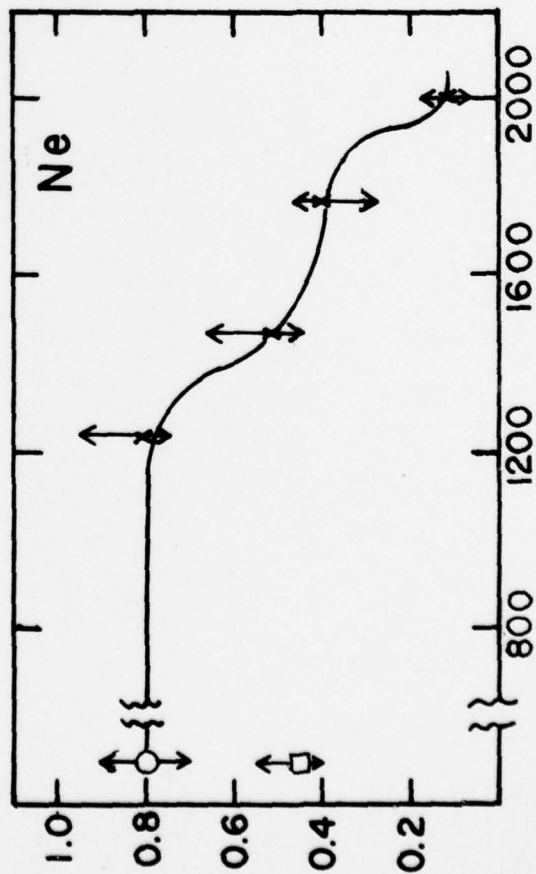
EAC



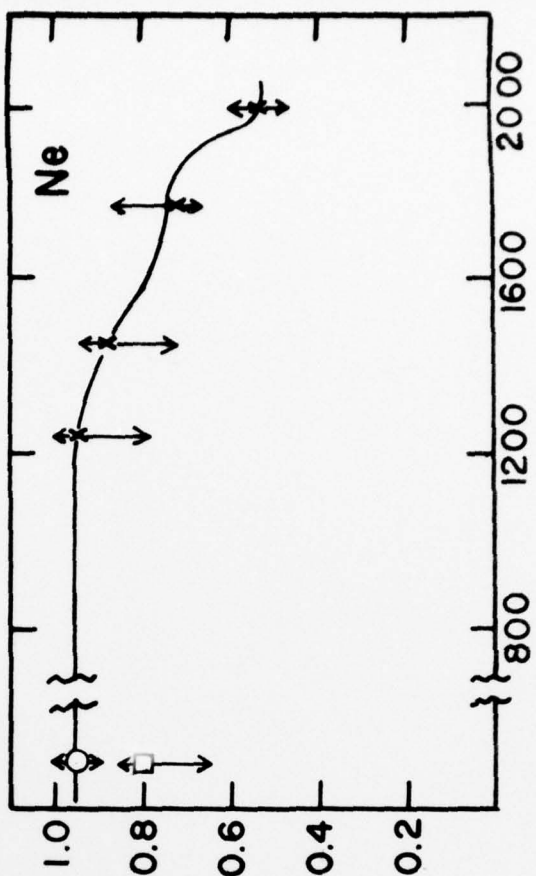
TMAC



Ne



Ne



flashing temp. ($^{\circ}\text{K}$)

Figure 5